EXCIMER LASER RESEARCH

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Semi Annual Report for Period 15 October 1976 to 15 March 1977

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This report describes an experimental investigation of the mercury mono-halides. As a class, these molecules show great promise for achieving high power scalable laser action in the visible portion of the spectrum.

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LIST OF ILLUSTRATIONS

I. INTRODUCTION

This Semi-Annual Technical Report covers the period 15 October 1976 through 15 March 1977 and describes the initial results of the new mercury monohalide lasers.

At present the demonstrated lasers which comprise the leading candidates to meet the ARPA need for an efficient, high-power laser capable of low loss atmospheric propagation include KrF,[1] XeF,[2] HgCl[3] and HgBr.[4] In the case of KrF, many important steps in the molecular formation and quenching kinetics have been measured and have provided the essential inputs for laser scaling designs. The present emphasis toward effective use of the KrF laser is the development of an efficient wavelength converter to avoid the sizable atmospheric absorption at the KrF wavelength, 2486 $\overset{o}{A}$. The present status of XeF laser development indicates that improvement of the laser efficiency is desirable to meet scaling goals. However, the XeF laser at 3540 $\overset{o}{A}$ provides an ideal wavelength for low-loss propagation. The most recently demonstrated lasers, HgCl and HgBr, operate at visible wavelengths 5576 $\overset{o}{A}$ and 5018 $\overset{o}{A}$, respectively, which will incur low propagation loss. Although initial HgCl laser measurements suggest high fluorescence efficiency, the formation and quenching kinetics of both $HgCl^{*}$ or $HgBr^{*}$ have yet to be determined.

(1) J.J. Ewing and C.A. Brau, Appl. Phys. Lett. __27__, 350 (1975).

(2) C.A. Brau and J.J. Ewing, Appl. Phys. Lett. __27__, 435 (1975).

(3) J.H. Parks, Appl. Phys. Lett. __31__, 192 (1977).

(4) J.H. Parks, Appl. Phys. Lett. __31__, 297 (1977).

II. MERCURY MONOHALIDE LASERS

In the last two years ARPA/ONR sponsored Visible/UV laser programs at AERL have led to the discovery and successful development of the rare gas halides. Specifically, we have obtained 12 J/liter with a 10% intrinsic efficiency from KrF[*] by pure e-beam pumping and 10 J/liter with a 9.5% intrinsic efficiency from e-beam controlled discharge pumping. In the last six months another class of visible lasers, the mercury halides, have been discovered. The technical papers describing these lasers are included in Appendix A. These molecules, like the rare gas halides, have ionic upper levels. Hence the formation kinetics of the upper laser level should be rapid and efficient. Because the ionization energy of mercury is lower than the rare gases, the mercury halides radiate at longer wavelengths than the rare gas halides. Table I lists the four mercury halides and their respective transition wavelengths.

A. HgX[*] LASER CHARACTERISTICS

This section summarizes the characteristics of the new high-power visible lasers operating on the $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ transition of HgCl at 5576 $\overset{o}{A}$ and the same molecular electronic transition of HgBr at 5018 $\overset{o}{A}$ and 4984 $\overset{o}{A}$. The upper laser state is ionic in nature and is formed directly by chemical reactions in an e-beam excited mixture of high pressure Ar/Xe and small amounts of Hg, CCl_4 and HBr. The lower laser level is the molecular ground state which is convalent in nature and bound by the order of 1 eV. The ionic character of the upper laser level provides the opportunity to utilize the highly efficient formation processes important in lasers such as KrF.

The lasing and spontaneous emission experiments were carried out in an aluminum cell, shown in Figures 1 and 2, in which the pressure could be varied up to pressures in the range of 7 atm. The cell temperature was controlled by cartridge heaters positioned to uniformly heat the gas cavity

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TABLE I

MERCURY HALIDE LASERS

Molecule	$\lambda \; \mathring{A}$	Lased
HgCl	5576	✓
HgBr	5018	✓
HgI	4450	
HgF	7000 - 9000	
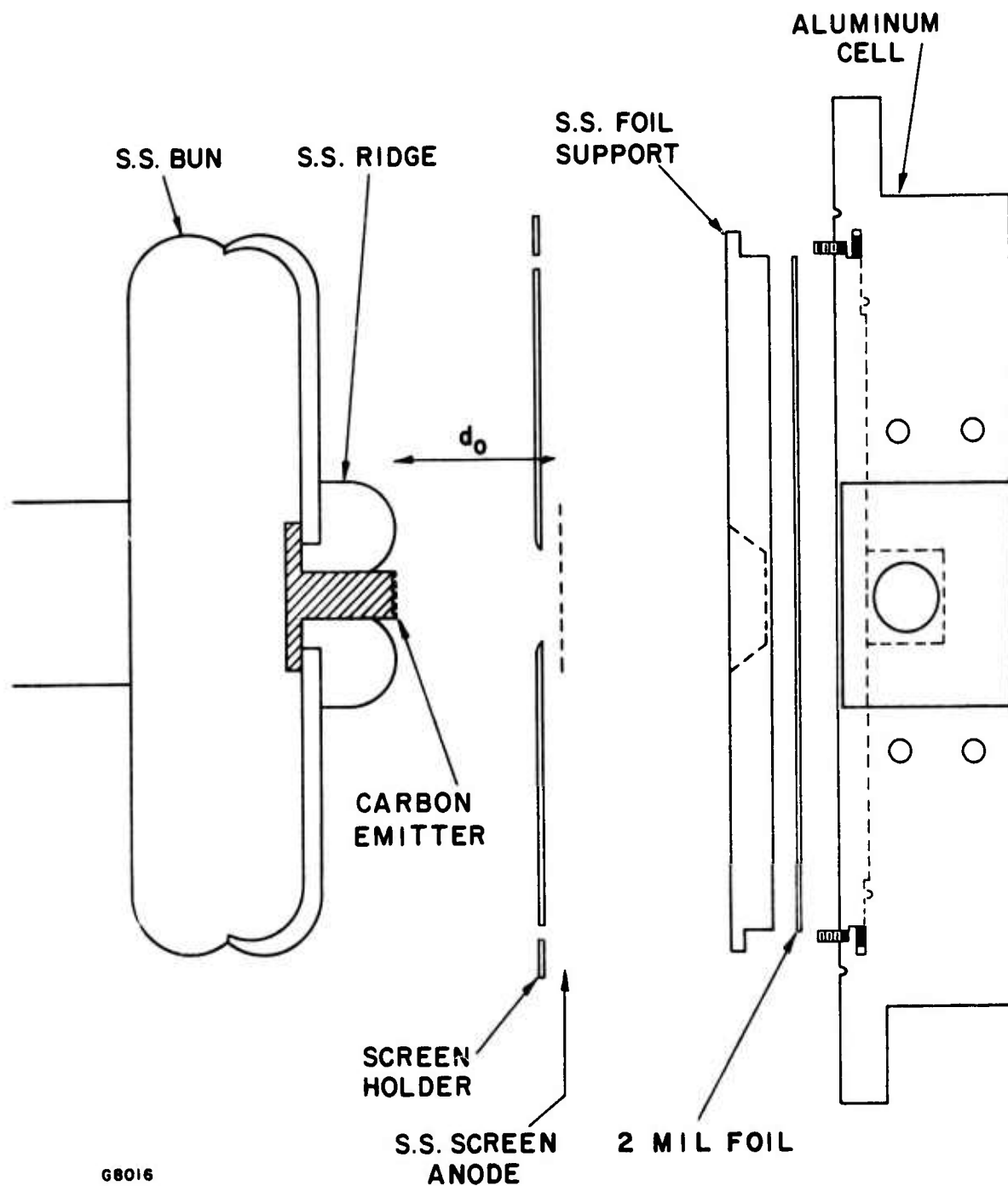
Figure 1   E-Beam/Laser Cell Configuration Indicating Transverse Beam
Excitation

CARTRIDGE
HEATERS

MIRROR
HOLDER

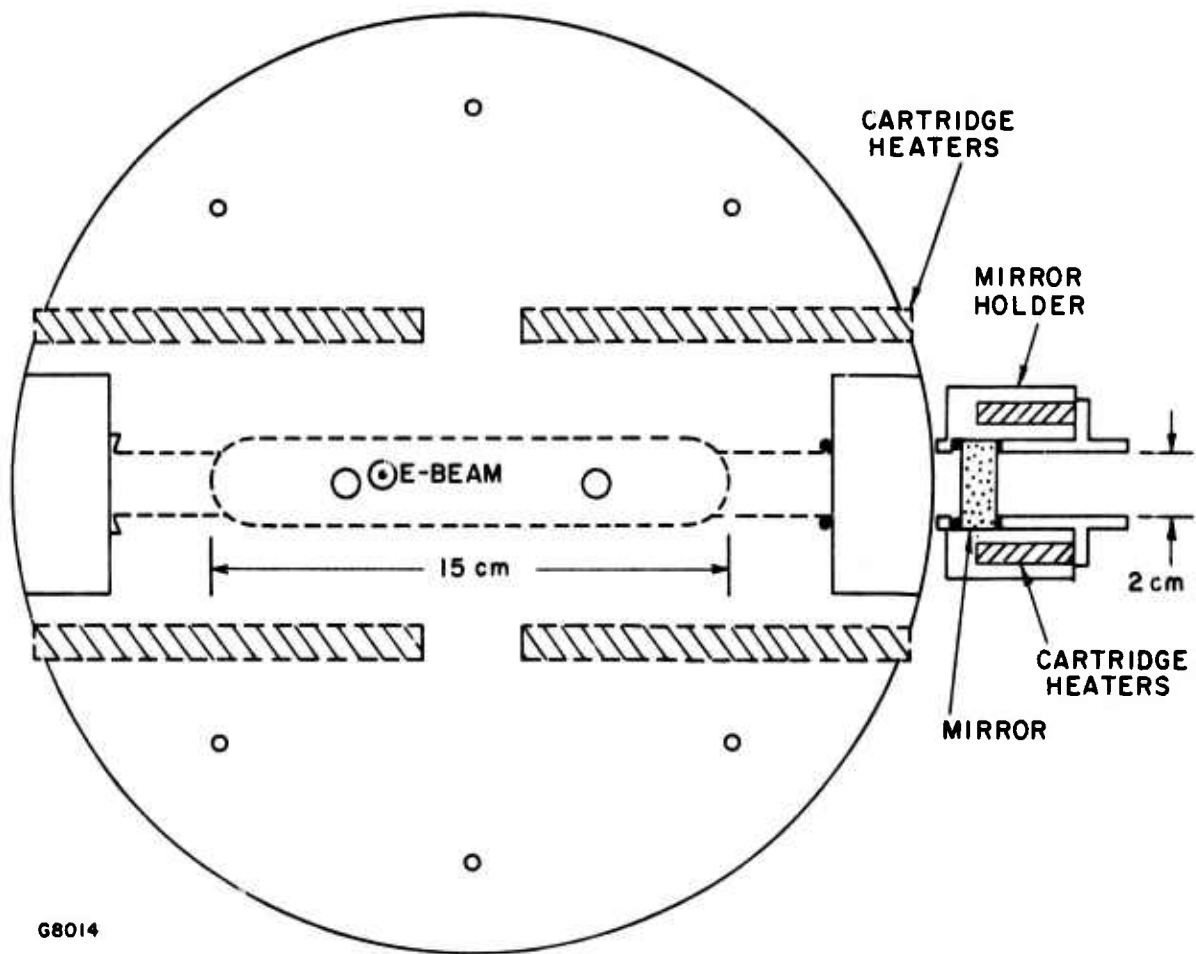E-BEAM

15 cm

2 cm

CARTRIDGE
HEATERS

MIRROR

G8014

Figure 2   Aluminum Laser Cell Showing Cartridge Heater Arrangement,
Cavity and Window Configuration

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and this temperature was kept below 300°C to allow the use of Viton O-rings. Laser mirrors are housed in heated aluminum holders providing an optical aperture of about 2 cm in diameter and sealed directly to the cell. The cavity mirrors are separated by 24 cm. A Marx generator is used to impulse charge a cold-cathode e-gun to about 300 kV for about 150 nsec providing a current density of about 100 A/cm^2 ir the cell. The e-beam of roughly 1 x 15 cm^2 cross section was injected into the gas transverse to the laser cavity optical axis through a 2-mil cell foil and irradiated a gas volume of about 85 cm^3. The e-beam energy deposition in the gas mixture was deduced from differential pressure measurements.

Laser action from HgCl was obtained in a typical mixture of Ar, Xe, Hg and CCl$_4$ in the ratio 85.7%/11.1%/2.1%/1.1% respectively at an Ar density of 3 amagats. The cell and Hg reservoir temperatures were 275°C and 260°C respectively. The laser output was viewed at one end with a calibrated planar photodiode (ITT F4000-S5) and at the other end with a 1/2 m Hilger quartz spectrograph. The HgCl spontaneous emission spectrum, shown in Figure 3, obtained in the presence of these high-pressure rare gas mixtures indicates that the most intense transitions occur between v' = 0 and high v'' ground state levels. The laser transition is identified as v' = 0 → v'' = 22 (HgCl^{35}) which corresponds to the strongest transition observed in spontaneous emission. The peak laser power obtained to date was about 1.7 MW with 50% output cooling as shown on the oscillogram trace in Figure 4(a). The associated pulse energy of 175 mJ corresponds to an intrinsic laser efficiency of about 3.5% considering the e-beam energy deposited in the laser volume was about 4.8 J. Bottlenecking of the v'' = 22 lower laser level is not limiting the laser pulse width of about 100 nsec. This has also been observed when the cavity flux was ~ 12 MW/cm^2 as shown in Figure 4(b) which implies a very rapid relaxation of the lower laser level of roughly ≤ 1 nsec.

Laser action from HgBr was obtained in a typical mixture of Ar, Xe, Hg and HBr in the ratio 86.4%/10.8%/2.0%/0.8% respectively at an Ar density of 3 amagats. The laser emission shown in Figure 5 is identified as the v' = 0 → v'' = 22 and v' = 0 → v'' = 21 bands which again correspond to the strongest transitions observed in spontaneous emission. The output coupling

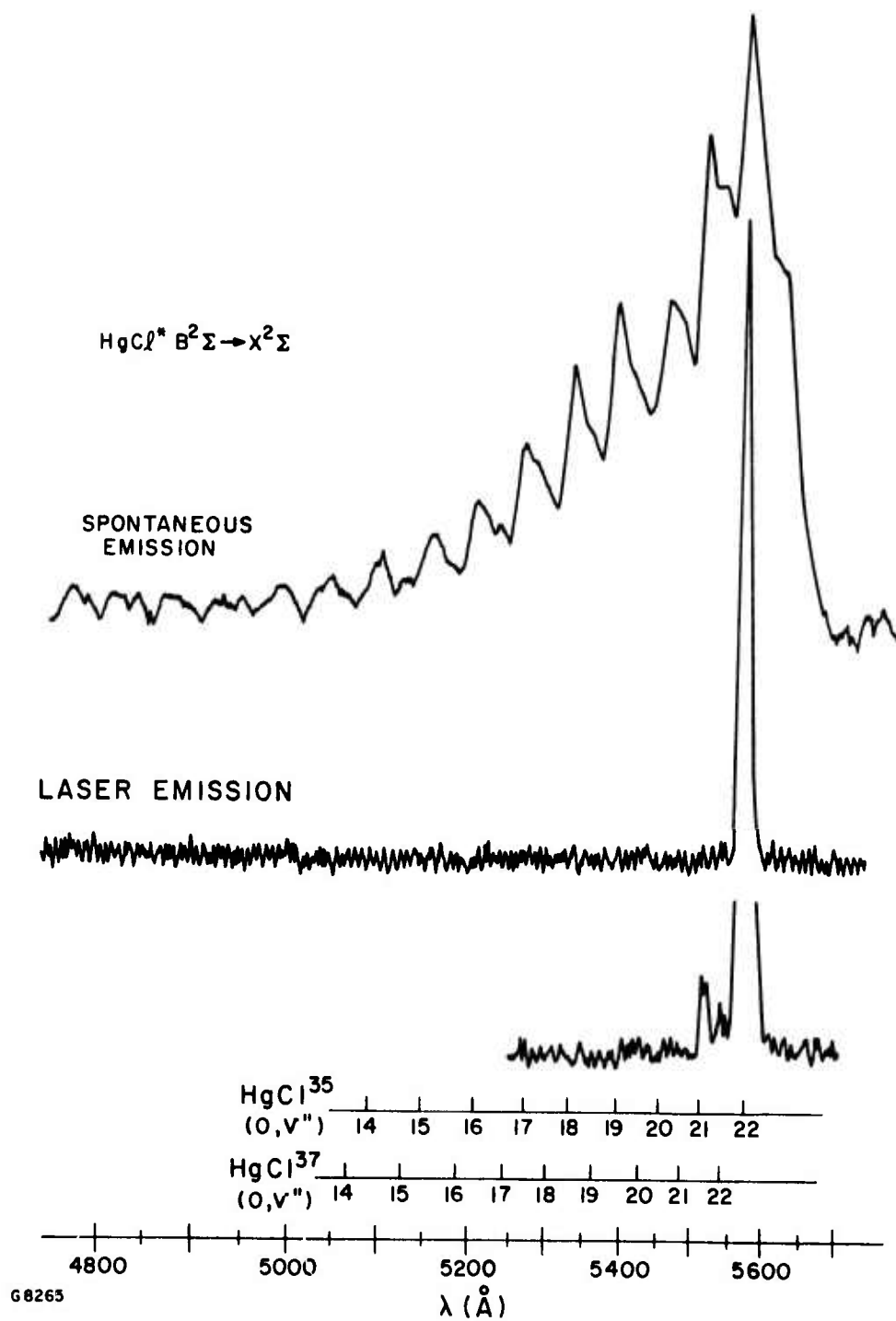Figure 3   Mercury Chloride Laser Emission and Spontaneous Emission
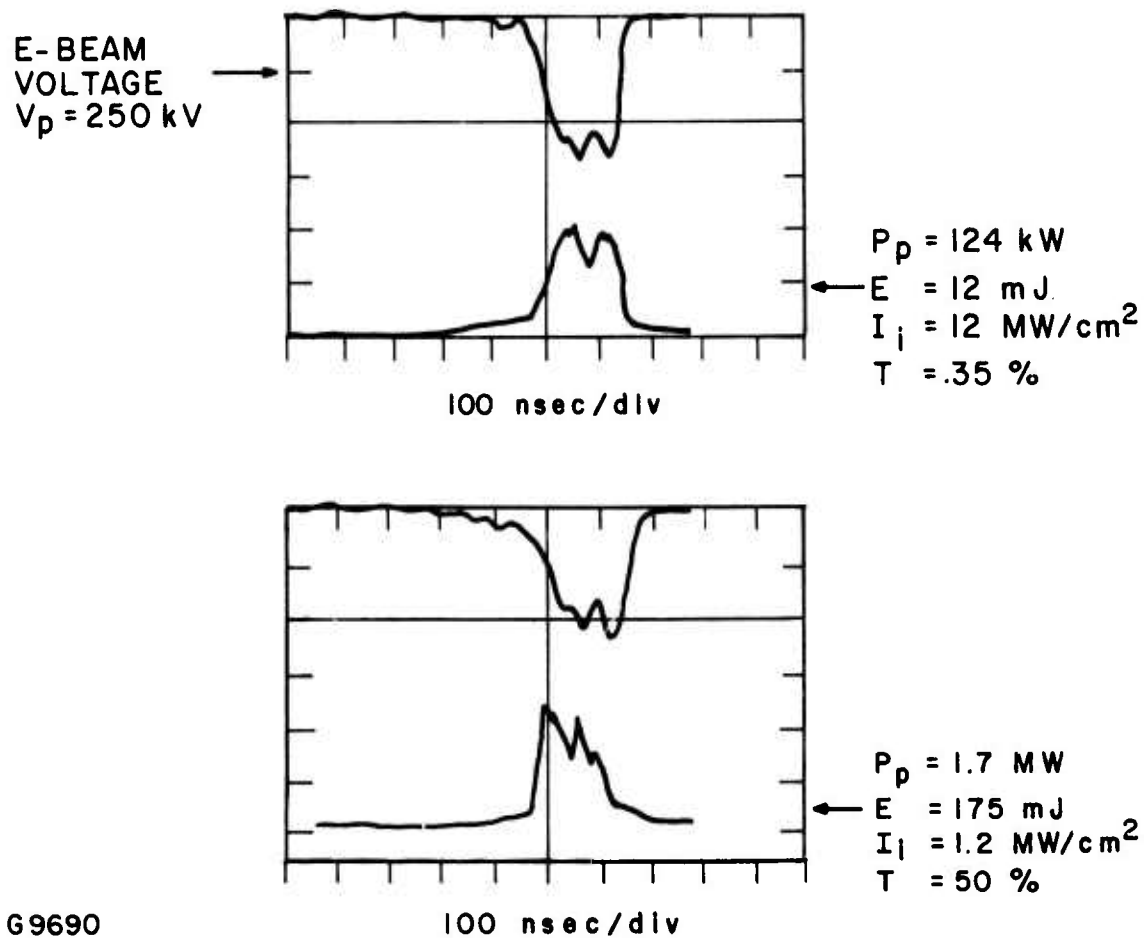   Spectra

E-BEAM
VOLTAGE
V_p = 250 kV

100 nsec/div

P_p = 124 kW
E = 12 mJ
I_i = 12 MW/cm^2
T = .35 %

P_p = 1.7 MW
E = 175 mJ
I_i = 1.2 MW/cm^2
T = 50 %

100 nsec/div

G9690

Figure 4   Oscillogram Trace Showing the E-Beam Voltage Pulse and the
Corresponding Photodiode Laser Signal for (a) Output Coupling
T = 50%; (b) Output Coupling T = .35%

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$HgBr^* B^2\Sigma \rightarrow X^2\Sigma$

SPONTANEOUS
EMISSION

LASER
EMISSION

Hg Br ^{79}
(O,V")   17 18 19 20 21 22

Hg Br ^{81}
(O,V")   17 18 19 20 21 22

4500   4600   4700   4800   4900   5000   5100   5200

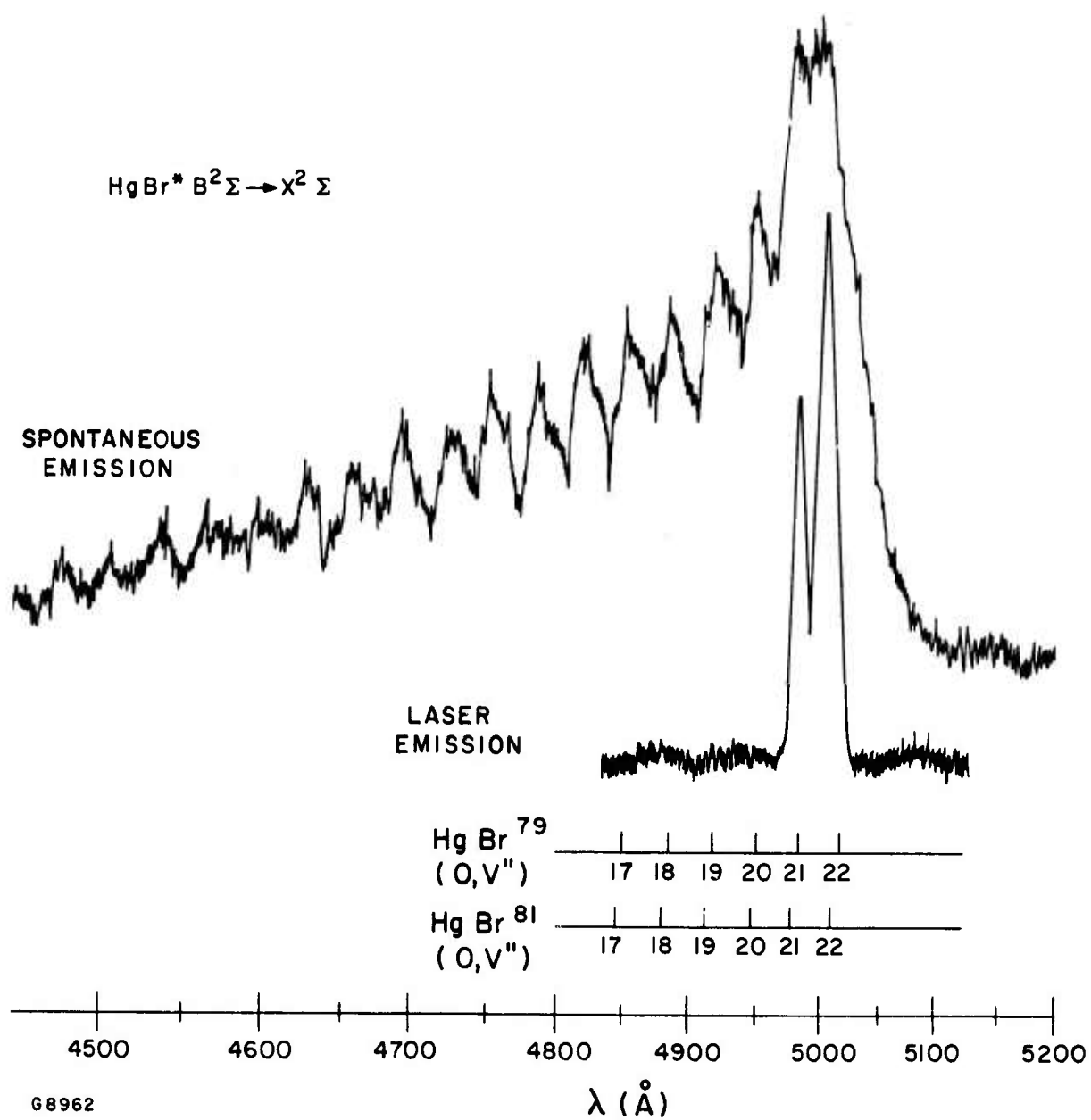G8962

λ (Å)

Figure 5   Mercury Bromide Laser Emission and Spontaneous Emission
            Spectra

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for each cavity mirror was 3%. The peak laser power of about 50 kW and integrated pulse energy of 3.2 mJ correspond to a laser efficiency of about 0.25%. The laser pulse width at half peak intensity is about 60 nsec and bottlenecking of the lower laser levels does not appear to be limiting the laser pulse length.

B. HgX* FORMATION KINETICS IN AN E-BEAM

A detailed study of the HgCl and HgBr molecular kinetics with e-beam excitation has not yet been performed. However, we will summarize the relative importance of several formation channels of HgCl* which are consistent with lasing and fluorescence data.

1. Ion-Ion Recombination

The dominant formation channel of HgCl* under our excitation conditions is the rapid three-body recombination of Hg^+ ions with Cl^- ions. The suggested ion-ion kinetic channeling is outlined in Figure 6. In gas mixtures containing high-pressure xenon, the high-energy beam electrons deposit most of their energy into the formation of Xe^+. The xenon ions and neutrals form Xe_2^+ with a three-body rate constant of $3.6 \times 10^{-31} cm^6/sec$.[5] The Xe_2^+ can then undergo charge transfer with Hg to form Hg^+ which is energetically near resonance, $\Delta E = 0.8$ eV (exothermic). This reaction may exhibit a comparable rate to an analogous, near resonant charge transfer reaction involving Ar_2^+ and Kr which forms Kr^+ with a rate constant of $7.5 \times 10^{-10} cm^3/sec$.[6] The Cl^- ion is formed by rapid dissociative attachment[7] of secondary electrons to CCl_4. The HgCl* ionic state is then formed by ion-ion recombination of Hg^+ and Cl^- which can have an effective two body rate constant of $10^{-6} cm^3/sec$ at pressure of interest.[8] In the Ar/Xe gas mixtures, the observed increase in HgCl* fluorescence is consistent with an

(5) D. Smith, A.G. Dean, and I.C. Plumb, J. Phys. B5, 2134 (1972).

(6) D.K. Bohme, N.G. Adams, M. Moselman, D.B. Dunkin and E.E. Ferguson, J. Chem. Phys. 52, 5094 (1970).

(7) L.G. Christophorou and J.A.D. Stockdale, J. Chem. Phys. 48, 1956 (1968). D. Spence and G.J. Schulz, J. Chem. Phys. 58, 1800 (1973).

(8) M.R. Flannery, in Case Studies in Atomic Collision Physics 2, edited by E.W. McDaniel and M.R.C. McDowell, North Holland, Amsterdam 1972, p. 3.
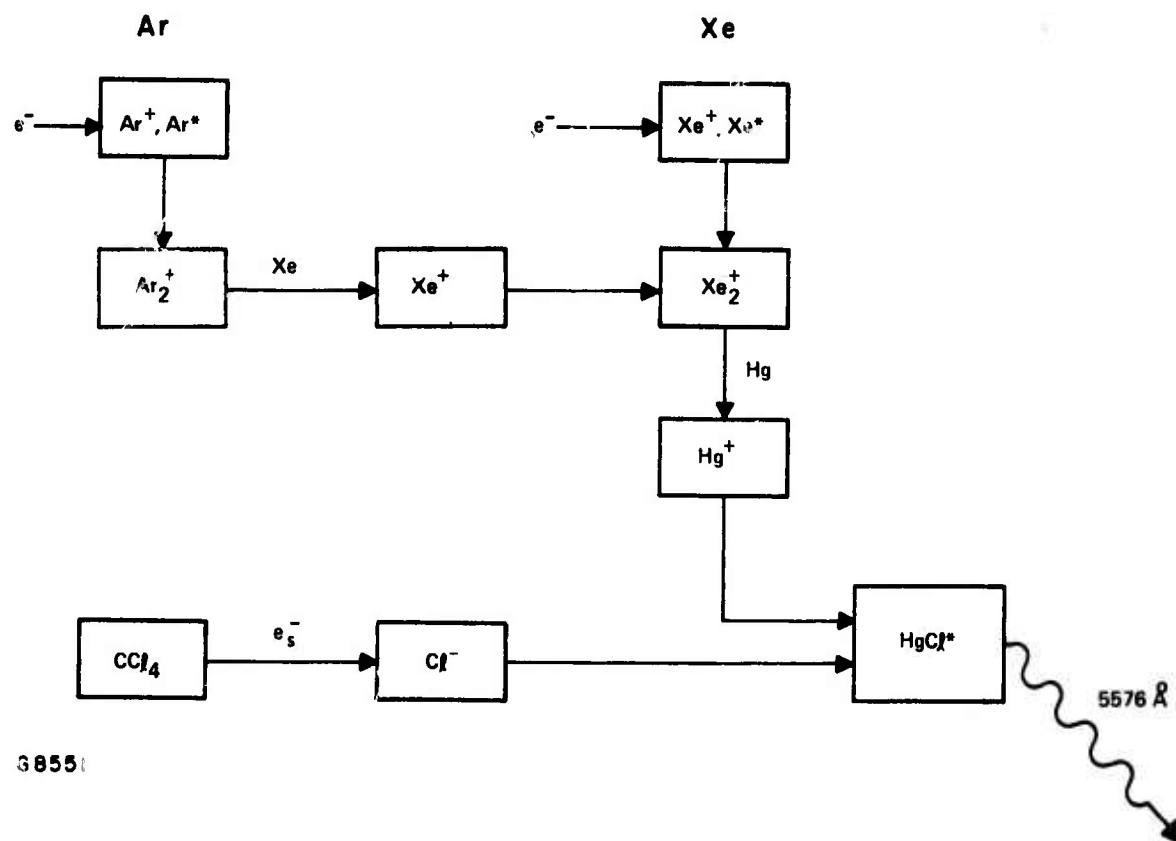
Figure 6   Ion-Ion Recombination Channel Suggested for HgCl* Formation in Ar/Xe/Hg/CCl$_4$ Mixtures

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enhanced formation of Hg^+. This could occur via additional charge transfer reactions involving Ar_2^+ and Xe which increase Xe_2^+ density and then Hg^+ as discussed above.

The absence of lasing in pure Ar mixtures and the observed weak lasing in pure Xe mixtures suggests the possibility that absorption[9] by Ar_2^* and Xe_2^* may be competing with $HgCl^*$ and $HgBr^*$ stimulated emission. The formation kinetics for these absorbing species is outlined in Figure 7.

2. Exchange Reactions and Interception

Fluorescence spectra obtained from $Ar/Hg/CCl_4$ and $Xe/Hg/CCl_4$ mixtures clearly indicate the formation of $ArCl^*$ and $XeCl^*$. Displacement reactions can follow in which mercury exchanges with these excited molecules to form $HgCl^*$ as shown in Figure 8. The rate constant for an analogous reaction involving Kr and ArF^* has been measured[10] to be 6.5×10^{-10} $cm^3/$sec. It is expected that $XeCl^*$ would be a more efficient precursor since dissociation to $Xe + Cl^*$ should have a smaller rate constant than the dissociation of $ArCl^*$ to $Ar + Cl^*$. Although the relative importance of this $HgCl^*$ formation channel has not been determined, the competing radiative processes of $ArCl^*$ and $XeCl^*$ suggest that displacement reactions will not be dominant.

The formation of a triatomic species, Kr_2F^* in KrF laser mixtures has been observed.[11] In this case, the triatomic formation is saturable since Kr_2F^* formation depends on the density of KrF^* which is driven down by the intense laser flux. Similar processes could be present in mercury halide laser mixtures. If Hg_2Cl^* is formed, it will probably fluoresce in the near infrared ≥ 7000 Å.

C. HgX^* FORMATION EFFICIENCY

Lasing on the $HgCl^*$ and $HgBr^*$ band transitions introduces new high-power laser sources at visible wavelengths. The detailed molecular kinetics in these gas mixtures have yet to be established in order to assess the possible efficiency and scalability of these lasers. The measured intrinsic efficiencies are the result of increased output coupling. The optimization of

(9) E. Zamir, D. L. Huestis, D. C. Lorents and H. H. Nakano, in Electronic Transition Lasers II, edited by L. E. Wilson, S. N. Suchard, and J. I. Steinfeld, MIT Press, Cambridge, Massachusetts 1977, p. 69.

(10) M. Rokni, J. H. Jacob, J. A. Mangano, and R. Brochu, Appl. Phys. Lett. 31, 79 (1977).

(11) J. A. Mangano, J. H. Jacob, M. Rokni, and A. Hawryluk, Appl. Phys. Lett. 31, 26 (1977).
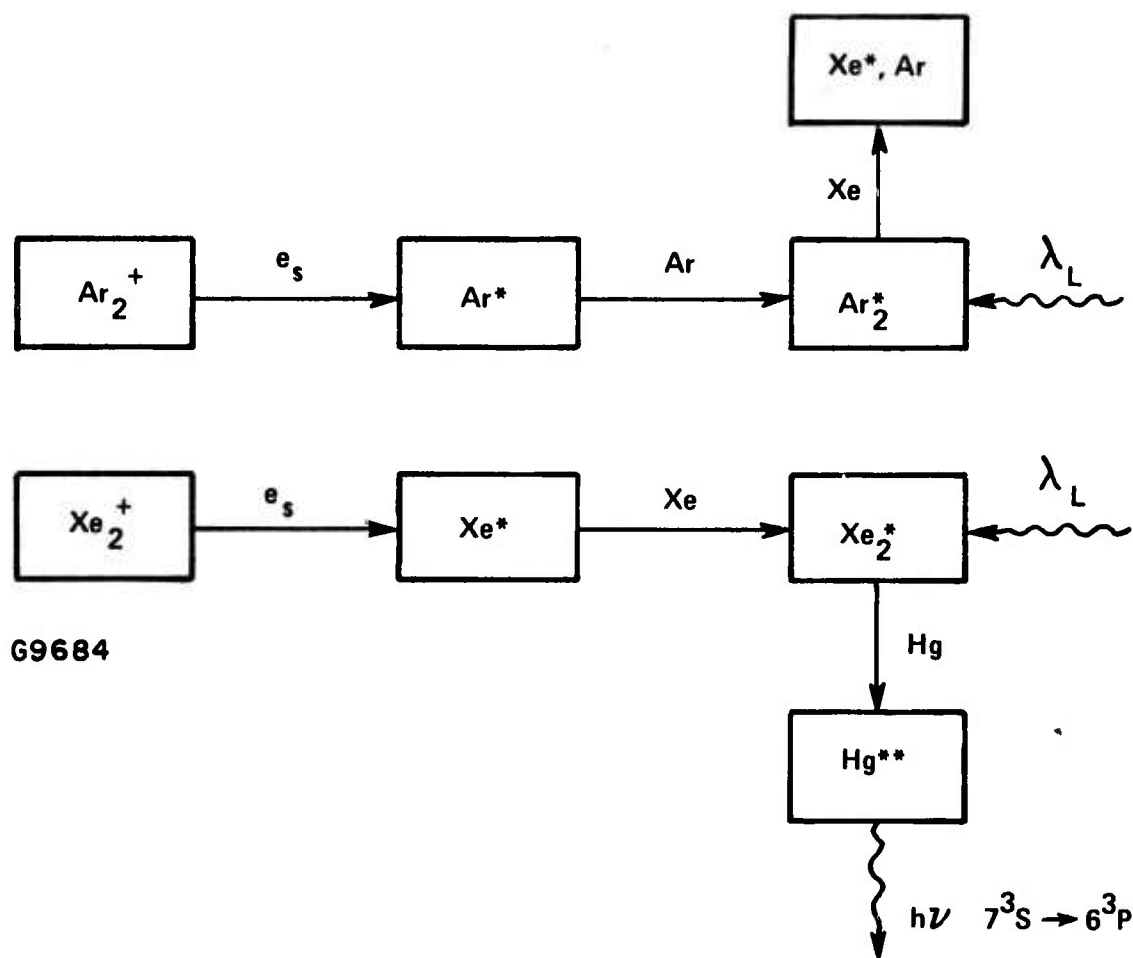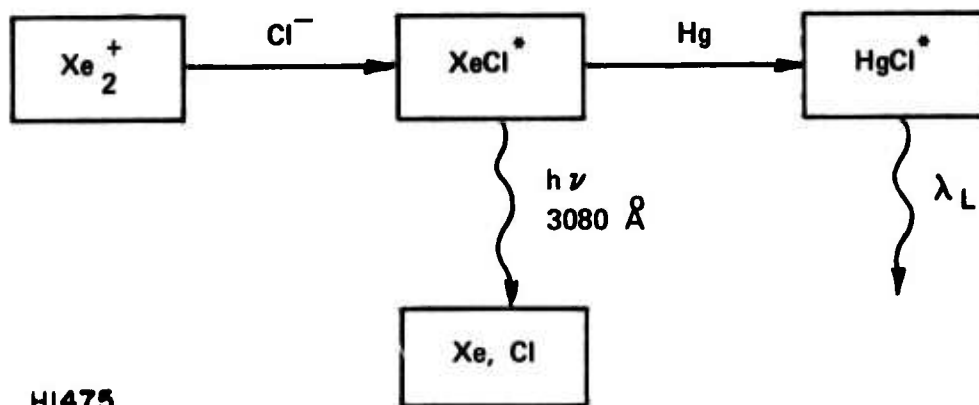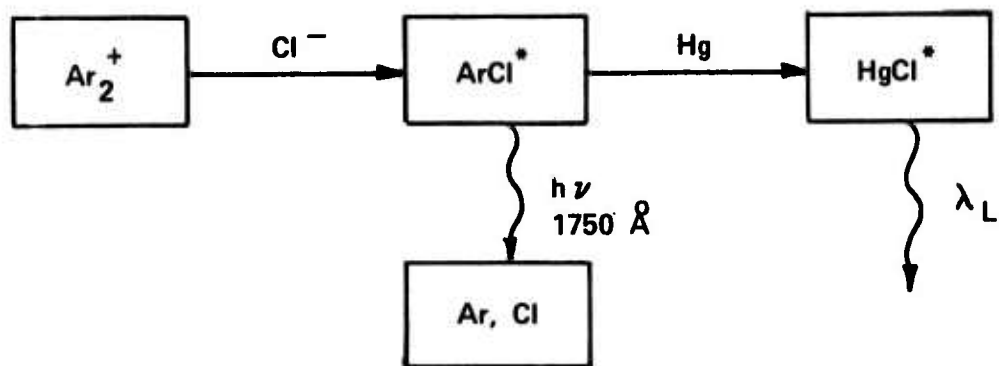
Figure 7   Suggested Formation Channels for Dominant Species which Absorb at the Laser Wavelength λ_L in $Ar/Xe/Hg/CCl_4$ Mixtures

Figure 8   HgCl* Formation via Exchange Reactions in Ar/Xe/Hg/CCl₄ Mixtures

laser mixtures and pumping power presently underway should result in higher values. In pure e-beam pumping, the maximum effective quantum efficiency of 9% is limited by the 26 eV needed to form an argon ion. Thus, the observed value of 3.8% indicates efficient $HgCl^*$ formation. If a discharge were used to pump the Hg^* (6p) metastable levels directly and form HgX^* via neutral reactions such as

$$Hg^* (^3P_2) + CCl_4 \rightarrow HgCl^* + CCl_3$$
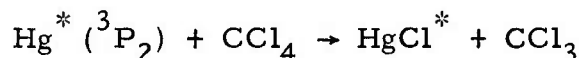
the effective quantum efficiency would be roughly 50%. The above reaction has been measured[12] to have a cross section of $34(\text{Å})^2$.

It was originally considered that halogen donor selection would be one of the more critical issues of discharge pumped lasing since the $Hg^* (^3P_0)$ metastable energy (~ 4.7 eV) is close to the average A-Cl bond strength (~ 3 eV). However, calculations[13] substantiated by measurements[14] in Ar-Hg discharges indicate that electron collisional mixing of the three metastables $Hg^* (^3P_{0,1,2})$ has a large enough cross section (~ 19 Å^2) to maintain the largest population in the highest $Hg^* (^3P_2)$ metastable at 5.43 eV. This considerably eases halogen donor selection and CCl_4 is an important candidate. In addition, this provides more flexibility to tailor the choice of halogen donor to the needs of discharge stability.

(12) H. F. Krause, S. G. Johnson, S. Datz, and F. K. Schmidt-Bleek, Chem. Phys. Lett. 31, 577 (1975).

(13) B. Yavorsky, Bull. Acad. Sci. U.S.S.R. 9, 233 (1945).

(14) C. Kenty, J. Appl. Phys. 21, 1309 (1950).

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7. L.G. Christophorou and J.A.D. Stockdale, J. Chem. Phys. $\underline{48}$, 1956 (1968). D. Spence and G.J. Schulz, J. Chem. Phys. $\underline{58}$, 1800 (1973).

8. M.R. Flannery, in Case Studies in Atomic Collision Physics 2, edited by E.W. McDaniel and M.R.C. McDowell, North Holland, Amsterdam 1972, p. 3.

9. E. Zamir, D.L. Huestis, D.C. Lorents and H.H. Nakano, in Electronic Transition Lasers II, edited by L.E. Wilson, S.N. Suchard, and J.I. Steinfeld, MIT Press, Cambridge, Massachusetts 1977, p. 69.

10. M. Rokni, J.H. Jacob, J.A. Mangano and R. Brochu, Appl. Phys. Lett. $\underline{31}$, 79 (1977).

11. J.A. Mangano, J.H. Jacob, M. Rokni and A. Hawryluk, Appl. Phys. Lett. $\underline{31}$, 26 (1977).

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13. B. Yavorsky, Bull. Acad. Sci. U.S.S.R. $\underline{9}$, 233 (1945).

14. C. Kenty, J. Appl. Phys. $\underline{21}$, 1309 (1950).

APPENDIX A

LASER ACTION ON THE $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ BAND OF HgCl AT 5576 Å

and

LASER ACTION ON THE $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ BAND OF HgBr AT 5018 Å

Laser action on the $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$ band of HgCl at 5576 Å[a)]

J. H. Parks

Avco Everett Research Laboratory, Inc., Everett, Massachusetts 02149

This letter describes a new laser operating on the $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$ band of HgCl at 5576 Å. Pumping was achieved by high-intensity electron-beam excitation of high-pressure rare-gas mixtures containing small amounts of Hg and CCl_4. A lower bound to the efficiency was determined to be 0.5% in an unoptimized configuration, and higher efficiency is expected.

PACS numbers: 42.55.Hq

This letter reports a new high-power visible laser operating on the $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$ transition of HgCl at 5576 Å ($v' = 0 \to v'' = 22$). The important spectroscopic features of HgCl have been observed previously.[1] This bound-bound laser transition originates on the first excited state of HgCl, $B^2\Sigma^+_{1/2}$, which is predominantly ionic in nature correlating with the separated ions $Hg^+(^2S_{1/2}) + Cl^-(^1S_0)$. This excited state is formed directly by chemical reactions in an e-beam-excited mixture of high-pressure rare gases and small amounts of Hg and CCl_4. It was found that a composition of Ar and about 10% Xe produced the most intense lasing conditions, as will be discussed below. The lower laser level is the molecular ground state $X^2\Sigma^+_{1/2}$ which is covalent in nature and bound by 1.04 eV. Although the detailed molecular parameters of HgCl derive from different electron configuration than the rare-gas monohalides, the ionic character of the upper laser level provides the opportunity to utilize the highly efficient formation processes important in lasers such as KrF.[2] For example, the production of HgCl* $B^2\Sigma$ via the harpoon reaction of $Hg^*(^3P_2)$ metastables with CCl_4 molecules, $Hg^* + CCl_4 \to HgCl^* + CCl_3$, has been observed[3] to have a reactive cross section of 34 Å^2 and a branching ratio into the upper laser level of near unity. Since the energies of the mercury metastables are about 5 eV, this reaction suggests the possibility of efficient discharge pumping of HgCl*.

The lasing and spontaneous emission experiments were carried out in an aluminum cell in which the pressure could be varied up to pressures in the range of 7 atm. The cell temperature was controlled by cartridge heaters positioned to uniformly heat the gas cavity and this temperature was kept below 300 °C to allow the use of Viton O-rings. Laser mirrors or quartz windows for fluorescence measurements are housed in heated aluminum holders providing an optical aperture of about 2 cm in diameter and sealed directly to the cell. A Marx generator is used to impulse charge a cold-cathode electron gun to about 300 kV for about 150 ns providing a current density of about 100 A/cm^2 in the cell. The e-beam, roughly 1×15 cm in area, was injected into the gas transverse to the laser cavity optical axis through a 2-mil aluminized Kapton foil and irradiated a gas volume of about 85 cm^3. The e-beam energy deposition in the gas mixture was deduced from differential pressure measurements.

Laser action from HgCl was obtained in a typical mixture of Ar, Xe, Hg, and CCl_4 in the ratio 85.7%/11.1%/2.1%/1.1%, respectively, at an Ar density of 3 amagats. The optimum Xe density was found to be about 0.4−0.5 amagat and increasing Xe to about 1.1 amagats decreases the laser intensity by a factor of 7. The cell and Hg reservoir temperatures were 275 and 260 °C, respectively. The cavity mirrors were separated by 24 cm and the cavity coupling out of each mirror was 0.35%. The laser output was viewed at one end with a calibrated planar photodiode (ITT F4000-S5) and at the other end with a $\frac{1}{2}$-m Hilger quartz spectrograph.

Figure 1 shows a comparison of the HgCl* spontaneous emission and laser spectra taken from microdensitom-
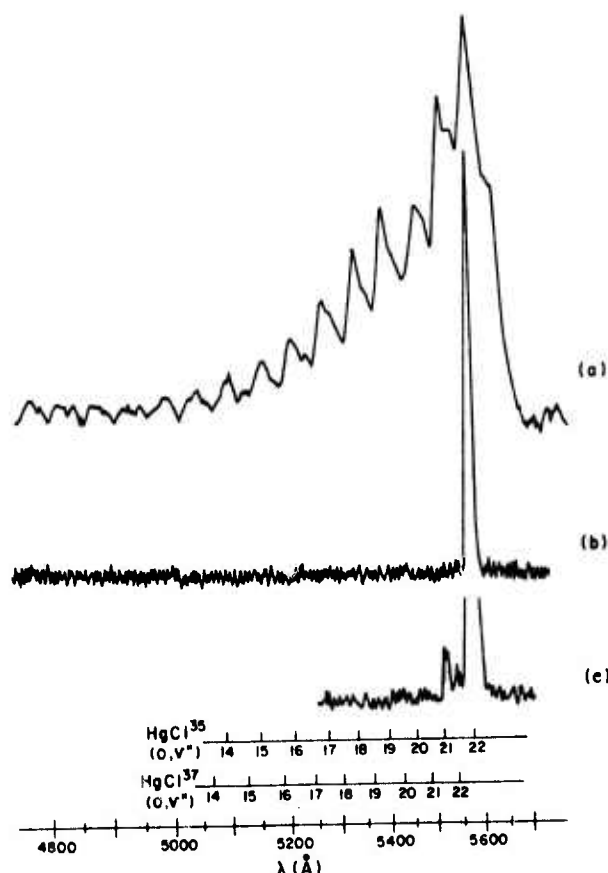

FIG. 1. A comparison of HgCl* fluorescence and laser spectra. (a) $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$ spontaneous emission spectra indicating the $v' = 0$ transitions, (b) the laser spectrum, and (c) exposure to several laser shots to observe weaker lasing transitions.
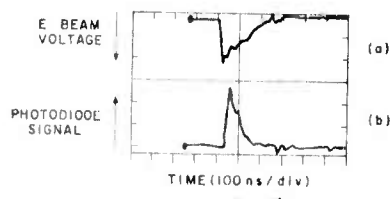
FIG. 2. Oscillogram trace showing (a) the e-beam voltage pulse and (b) the corresponding photodiode laser signal.

eter traces of Kodak 103-F plates. Figure 1(a) shows the $B \rightarrow X$ spontaneous emission spectrum obtained in the presence of these high-pressure rare-gas mixtures. The most intense transitions occur between $v' = 0$ and high v'' ground-state levels as indicated, although several of the weaker transitions originate on $v' = 1$. The laser transition shown in Fig. 1(b) is identified as $v' = 0 \rightarrow v'' = 22$ (HgCl^{35}) which corresponds to the strongest transition observed in spontaneous emission. Exposure of the plate to several laser pulses shown in Fig. 1(c) reveals two additional laser transitions, many orders of magnitude weaker, which are identified as $v' = 0 \rightarrow v'' = 21$ (HgCl^{35}) and $v' = 0 \rightarrow v'' = 22$ (HgCl^{37}). Since the laser emission heavily saturated the photographic plates, Figs. 1(b) and 1(c) do not reliably represent the laser spectral linewidths.

Figure 2 shows an oscillogram of the photodiode signal from the HgCl laser in trace (b) and the e-beam voltage pulse monitor in trace (a). The laser intensity was attenuated by a factor 2.6×10^{-6} before reaching the photodiode and a narrow-band filter isolating the 5576-Å band transition was placed between the neutral density attenuators and the photodiode surface. The e-beam voltage pulse has a peak value of 280 kV. The photodiode signal corresponds to a peak laser power of about 138 kW and an integrated pulse energy of 8 mJ. The energy deposited by the e-beam in the irradiated gas volume was deduced to be 6 J. Considering that the lasing volume fills about 0.5 of the irradiated gas volume, the total laser output from both ends of 16 mJ corresponds to a laser efficiency of about 0.5%. Since the loss introduced by the laser mirrors is comparable to the output coupling, the efficiency may be closer to 1%. This is certainly a lower bound and optimization of laser mixtures, pumping power, and output coupling should result in higher efficiency. The laser pulse width at half peak intensity is about 40 ns, and broadens to 120 ns near the base of the pulse which is comparable with the pulse width observed in spontaneous emission. Bottlenecking of the $v'' = 22$ lower laser level does not appear to be limiting the laser pulse length. The product of the HgCl* stimulated emission cross section, σ_s, and lifetime τ is estimated from the observed spontaneous emission spectrum to be about 1×10^{-23} cm^2 sec. The saturation flux in the absence of HgCl* quenching is $\phi_s = h\nu/\sigma_s\tau \sim 35$ kW/cm^2 which is much smaller than the peak internal cavity flux of ~ 14 MW/cm^2 inferred from the laser output power. This implies that relaxation of the high-lying $X^2\Sigma$ vibrational levels is sufficiently rapid at these high pressures to prevent a lower level bottleneck in the presence of this saturating laser flux. The level spacing at $v'' = 22$ is

about 0.6 kT_{gas} at the cell temperature of 548 K. In addition, the $v'' = 22$ level is about 0.7 eV above the $v'' = 0$ level, which would result in a small population of the lower laser level even in the presence of such rapid relaxation.

Laser action was also obtained in mixtures of Xe, Hg, and CCl$_4$ typically in the ratio 91%/5.8%/3.2%, respectively, at a Xe density of 1.1 amagat. The cell and Hg reservoir temperature was 275 and 260 C, respectively. Although a complete parametric pressure dependence has not been performed, lasing was not observed for Xe density below 0.4 amagat. The peak laser intensity using pure Xe was down about a factor of 3 from Ar/Xe gas mixtures. A comparable decrease was observed in HgCl* fluorescence although the relative e-beam energy deposited in the Xe and Ar/Xe mixtures mentioned above was 4.5 and 6 J, respectively. Laser action was not obtained when pure Ar was used as the high-pressure component, although comparable HgCl* fluorescence was observed and the e-beam energy deposited in Ar was 4.5 J.

A detailed study of the HgCl* molecular kinetics with e-beam excitation has not been performed. However, the dominant formation channel of HgCl* under our excitation conditions and consistent with lasing and fluorescence data is the rapid three-body recombination of Hg$^+$ ions with Cl$^-$ ions. In Xe/Hg/CCl$_4$ mixtures, the primary electrons deposit most of their energy into the formation of Xe$^+$ which rapidly forms[1] Xe$_2^+$. The Xe$_2^+$ can then undergo near-resonant charge transfer with Hg to form Hg$^+$. This reaction may exhibit a comparable rate to the analogous reaction involving Ar$_2^+$ and Kr which forms Kr$^+$ with a rate constant of 7.5×10^{-10} cm^3/sec.[5] The Cl$^-$ ion is formed by rapid dissociative attachment[6] of secondary electrons to CCl$_4$. The HgCl* ionic state is then formed by ion-ion recombination of Hg$^+$ and Cl$^-$ which can have an effective two-body rate constant of $\sim 10^{-6}$ cm^3/sec at pressures of interest.[7] In the Ar/Xe gas mixtures, the observed increase in HgCl* fluorescence is consistent with an enhanced formation of Hg$^+$.

The mercury metastable channel leading to HgCl* via harpoon reactions is probably not an important formation process under our excitation conditions. The absence of strong Hg neutral transitions indicates that mercury metastable production is inefficient in these laser gas mixtures. Fluorescence spectra obtained from Ar/Hg/CCl$_4$ and Xe/Hg/CCl$_4$ mixtures clearly indicate the formation of ArCl* and XeCl*. Displacement reactions[8] can follow in which mercury exchanges with these excited molecules to form HgCl*. However, the competing radiative processes of ArCl* and XeCl* suggest that these reactions will not be dominant.

The absence of lasing in pure Ar mixtures and the observed weak lasing in pure Xe mixtures suggests the possibility that absorption[9] by Ar$_2^+$ and Xe$_2^+$ may be competing with the HgCl* stimulated emission. The fluorescence from pure Ar and pure Xe mixtures was comparable for similar e-beam energy deposition.

However, in Ar/Xe/Hg/CCl$_4$ gas mixtures, the strong Ar$_2^*$ absorption is probably reduced via the rapid quenching[10,11] of Ar$_2^*$ by Xe.

In summary, lasing on the HgCl* band transition at 5576 Å has been attained and introduces a new high-power laser source at a visible wavelength. The detailed molecular kinetics in Ar/Xe/Hg/CCl$_4$ mixtures have yet to be established in order to assess the possible efficiency and scalability of this laser. In pure e-beam pumping, the effective quantum efficiency of 8.6% is limited by the 26 eV needed to form an argon ion. If a discharge were used to pump the Hg* (6p) metastable levels directly, the effective quantum efficiency would be roughly 45%. Of the mercury halides, HgCl offers the possibility of having the highest gain because of its longer wavelength and lowest intrinsic absorption loss. Laser action on the $^2\Sigma \rightarrow {}^2\Sigma$ transition of HgBr and HgI is possible; however, the shorter wavelengths of these transitions may result in greater self-absorption losses.

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Laser action on the $B^2\Sigma^+_{1/2}\to X^2\Sigma^+_{1/2}$ band of HgBr at 5018 Å[a)]

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This letter describes a new laser operating on the $B^2\Sigma^+_{1/2}\to X^2\Sigma^+_{1/2}$ band of HgBr at 5018 Å. Pumping was achieved by high-intensity electron-beam excitation of high-pressure Ar/Xe mixtures containing small amounts of Hg and HBr. A lower bound to the efficiency was determined to be 0.25% in an unoptimized configuration, and higher efficiency is expected.

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Recently we reported[1] laser action on the $B^2\Sigma - X^2\Sigma$ band of HgCl at 5576 Å. This letter reports laser action on the corresponding band in another member of this class of molecules, HgBr at 5018 Å. The important spectroscopic features of HgBr which have been observed previously[2,3] indicate that this bound-bound laser

transition originates on the excited ionic state $B^2\Sigma^+_{1/2}$. The molecule is formed directly in this state by chemical reactions in an e-beam-excited mixture of high-pressure rare gases and small amounts of Hg and HBr. The lower laser level is the molecular ground state $X^2\Sigma^+_{1/2}$ which is covalent in nature and bound by about 0.7 eV. As emphasized previously for HgCl*, the ionic character of the upper laser level provides the opportunity to take advantage of highly efficient excited-state-formation processes. For example, harpooning reactions of mercury metastables and molecules contain-

ing bromine atoms should rapidly form HgBr* molecules in the upper laser level as has been observed[4] for HgCl. Since the relevant energy levels of mercury metastables only require about 5 eV, the formation of HgBr* via this reaction suggests the possibility of efficient discharge pumped lasing.

The aluminum cell used in these lasing and spontaneous-emission experiments has been described previously.[1] A Marx generator is used to impulse charge a cold-cathode electron gun to about 300 kV for about 150 ns providing a current density of about 100 A/cm^2 in the cell. The e-beam of roughly 1×15 cm^2 cross section was injected into the gas transverse to the laser cavity optical axis through a 2-mil aluminized Kapton foil and irradiated a gas volume of about 85 cm^3. The e-beam energy deposition in the gas mixture was deduced from differential pressure measurements before and after irradiation.

Laser action from HgBr was obtained in a typical mixture of Ar, Xe, Hg, and HBr in the ratio 86.4%/10.8%/2.0%/0.8%, respectively, at an Ar density of 3 amagats. The cell and Hg reservoir temperatures were 275 and 260 °C, respectively, and the rare gases were preheated. The laser cavity mirrors, sealed directly to the cell by Viton O-rings, were separated by 24 cm and had optical transmissions of 2.5 and 3% at the laser wavelength. The laser output was viewed at one end with a calibrated planar photodiode (ITT F4000-S5) and at the other end with a $\frac{1}{2}$-m Hilger quartz spectrograph. Figure 1 shows a comparison of the HgBr* spontaneous-emission and laser spectra taken from microdensitometer traces of Kodak 103-F plates. Figure 1(a) shows the $B \to X$ spontaneous-emission

spectrum obtained in the presence of these high-pressure rare-gas mixtures. The most intense transitions occur between $v'=0$ and high v'' ground-state levels as indicated, although several of the weaker transitions originate on $v'=1$. The mercury neutral atom transitions at 4358 and 5461 Å are observed to the left and right of the HgBr* bands, respectively. The laser emission shown in Fig. 1(b) is identified as the $v'=0 \to v''=22$ and $v'=0 \to v''=21$ bands which correspond to the strongest transitions observed in spontaneous emission. Since the isotopic abundance of Br^{79} and Br^{81} is 50.54 and 49.46%, respectively, and the spectral resolution is not adequate to resolve the isotope shift of ~6 Å, it is assumed that the laser emission includes band transitions of both isotopic molecular species. Since the laser emission heavily saturated the photographic plates, Fig. 1(b) does not reliably represent the laser spectral linewidths.

Figure 2 shows an oscillogram of the photodiode signal from the HgBr laser in trace (b) and the e-beam voltage pulse monitor in trace (a). The laser intensity was attenuated by a factor of 5×10^4 before reaching the photodiode and a bandpass filter peaked about 5000 Å was placed between the neutral density attenuators and the photodiode surface. The e-beam voltage pulse has a peak value of 240 kV. The photodiode signal corresponds to a peak laser power of about 50 kW and an integrated pulse energy of 3.2 mJ. The energy deposited by the e-beam in the irradiated gas volume was deduced to be 6 J. Considering that the lasing volume fills about 0.5 of the irradiated gas volume, the total laser output from both ends of 7 mJ corresponds to an intrinsic laser efficiency of about 0.25%. This is certainly a lower
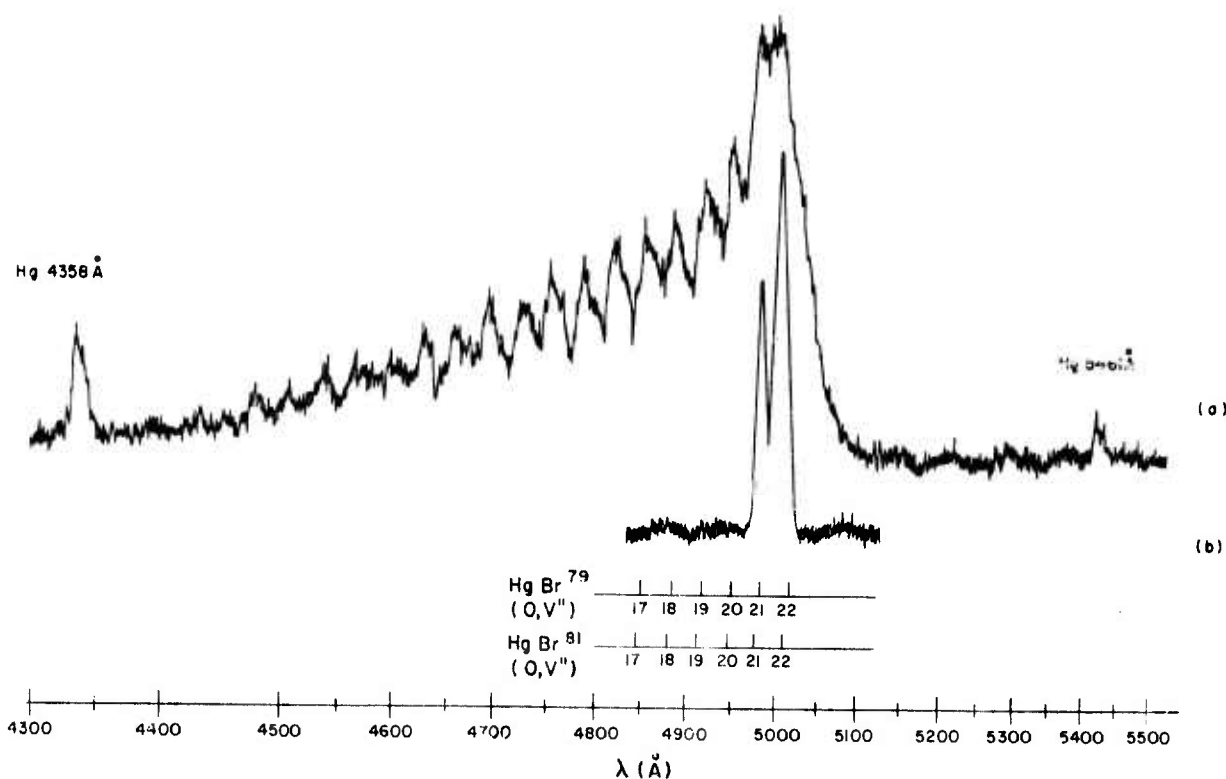

FIG. 1. A comparison of HgBr* fluorescence and laser spectra. (a) $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$ spontaneous-emission spectra indicating the $v'=0$ transitions and several mercury lines; (b) the laser spectrum.
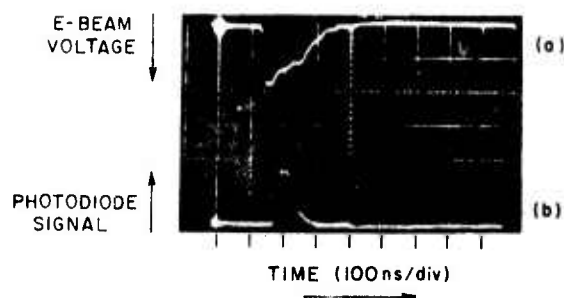
FIG. 2. Oscillogram trace showing (a) the e-beam voltage pulse and (b) the corresponding photodiode laser signal.

bound, and optimization of laser mixtures, pumping power, and output coupling should result in higher efficiency. The laser pulse width at half-peak intensity is about 60 ns and broadens to 130 ns near the base of the pulse which is comparable with the pulse width observed in spontaneous emission. Bottlenecking of the lower laser levels does not appear to be limiting the laser pulse length. The product of the HgBr* stimulated-emission cross section σ_s and lifetime τ is estimated from the observed spontaneous-emission spectrum to be about 5.6×10^{-24} cm^2 s. From the measured[5] value of the $B \rightarrow X$ radiative lifetime, $\tau = 23.3$ ns, the cross section is estimated to be $\sigma_s = 2.4 \times 10^{-16}$ cm^2. The saturation flux in the absence of HgBr* quenching is $\phi_s = h\nu/\sigma_s\tau \sim 70$ kW/cm^2 which is an order of magnitude less than the peak internal cavity flux of ~ 0.7 mW/cm^2 inferred from the laser output power. The level spacing at $v'' = 22$ is about $0.5 k T_{gas}$ at the cell temperature of 548 °K. In addition, the $v'' = 22$ level is about 0.45 eV above the $v'' = 0$ level, which would result in a small thermal population of the lower laser level even in the presence of rapid relaxation of the $X^2\Sigma$ vibrational levels.

Laser action was also obtained in mixtures of Xe, Hg, and HgBr typically in the ratio 92.5%/5.9%/1.6%, respectively, at a Xe density of 1.1 amagat. The peak laser intensity and pulse energy using pure Xe were down about a factor of 10 from Ar/Xe gas mixtures, although the HgBr* fluorescence intensities from pure Xe and Ar/Xe mixtures were comparable. The relative e-beam deposition in Xe and Ar/Xe was 4.5 and 6 J, respectively. Laser action was not obtained when pure Ar was used as the high-pressure component.

A detailed study of the HgBr* molecular kinetics with e-beam excitation has not been performed. However, we will consider several formation channels of HgBr* which are consistent with lasing and fluorescence data. The dominant formation channel of HgBr* under our excitation conditions appears to be the rapid three-body recombination of Hg$^+$ ions with Br$^-$ ions. The kinetic sequence involved in this formation channel are similar to those previously discussed[1] for HgCl*. However, HgBr* does not appear to form as efficiently as HgCl*, as evidenced by generally weaker fluorescence intensities for comparable e-beam energy deposition. This may result from the different intrinsic properties of the halogen donor molecules. HBr has a smaller dissociative attachment cross section[6,7] than CCl$_4$ by about a factor of 60 and is peaked at 0.5 eV, whereas the CCl$_4$

cross section is peaked near 0.02 eV. As a result, electron dissociative recombination[8] of Xe$_2^+$, $e + Xe_2^+$ $\rightarrow$ Xe** + Xe producing highly excited xenon atoms, is a competing process in HBr mixtures under our discharge conditions. This has the effect of reducing the formation of Br$^-$, and also Hg$^+$ which can result from near resonant charge exchange with Xe$_2^+$. The products of this dissociative recombination channel[9] into xenon atomic metastables and also Xe$_2^*$ molecules. It has been suggested that near-resonant energy transfer collisions from these neutral xenon species to 7^{3}S mercury levels result in the intense 7^{3}S mercury transitions observed in Xe/Hg mixtures.[9] Thus, the 7^{3}S mercury transitions at 4358 and 5461 Å, which we observe to be more intense in mixtures containing HBr than CCl$_4$, provide evidence that the dissociative recombination of Xe$_2^+$ can be a competing process depending on the halogen donor and its concentration.

The mercury metastable channel leading to HgBr* via a harpoon reaction is not an important formation process for this bromine donor. The reaction of HBr with all 6^{3}P mercury metastables is endothermic by $\geqslant 1.7$ eV. A more useful donor for this formation channel might be CBr$_4$ which has a bromine bond strength of about 2.4 eV.

Fluorescence spectra obtained from these gas mixtures indicate the formation of XeBr* which emits near 2818 Å. Displacement reactions can follow in which mercury exchanges with these excited molecules to form HgBr*. However, the competing XeBr* spontaneous-emission channel may reduce the importance of this formation process.

The absence of lasing in pure Ar mixtures and the observed weak lasing in pure Xe mixtures suggest the possibility that absorption by Ar$_2^*$ and Xe$_2^*$ excimers[10] may be competing with the HgBr* stimulated emission. As noted above, the dissociative recombination of Xe$_2^+$ will increase the density of Xe$_2^*$ molecules. However, in Ar/Xe/Hg/HBr gas mixtures, the Ar$_2^*$ absorption is possibly reduced via the rapid quenching[11,12] of Ar$_2^*$ by Xe and, in addition, the smaller density of Xe reduces the Xe$_2^*$ absorption.

In summary, lasing on the HgBr* band transition at 5018 Å has been attained and becomes the second member of the mercury halides to demonstrate lasing. The detailed molecular kinetics in Ar/Xe/Hg/HBr mixtures have yet to be established in order to assess the possible efficiency and scalability of this laser. In pure e-beam pumping, the effective quantum efficiency of 9.5% is limited by the 26 eV needed to form an argon ion. If discharge were used to pump the Hg*(6p) metastable levels directly, the effective quantum efficiency would be about 50%. Thus, HgBr and also HgCl show promise for higher efficiencies than have been previously achieved for visible lasers.

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